Air pollution from residential wood combustion in a Danish village

Indoor-outdoor measurements

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Contents

1 Introduction

The present note describes work conducted as part of a comprehensive research project entitled Residential Wood Combustion and the interaction between technology, user and environment – in short: WOODUSE. The project is interdisciplinary and covers many aspects of wood smoke pollution, as it investigates emissions, ambient and indoor pollution levels, health effects and human exposure, social aspects and abatement options.

Within the Wooduse project a three-month measuring campaign was conducted in the Danish village of Slagslunde, targeted at the study of residential wood combustion (Wåhlin et al., 2010). A sub-campaign was conducted during two weeks in order to examine the relation between indoor and outdoor pollution in two homes: one with, and the other without a woodstove.

The present note is concerned with these indoor-outdoor measurements; in particular it reports on measurements of transport of air, both in respect to outdoor-to-indoor transport, as well as between-room transport.

The note first gives a brief review of various results on indoor-outdoor air pollution derived from literature. Next, it goes on to describe flow and concentration measurements from the indoor-outdoor component of the Slagslunde measuring campaign.

Besides the present note many other publications have been produced within the Wooduse project; the project web site http://wooduse.dmu.dk gives an overview of all published material.

Other reports including information on the indoor-outdoor campaign are those of Wåhlin et al. (2010) and a report in Danish by Olesen et al. (2010).

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2 Literature review

2.1 Outdoor to indoor transport of airborne particles

As documented in several repeated surveys, humans typically spend up to 90% of their time in different indoor settings (e.g. Jenkins et al. 1992, Klepeis et al. 2001). Based on telephone interviews with over 9000 respondents across the US, Klepeis et al. (2001) reported an average of 83% of this time being spent in enclosed buildings and 69% in the home. Even with the considerable individual differences between humans and their behavior and activities, it is clear that the indoor air in residences represents a significant part of the overall environmental exposure. Nevertheless, epidemiological studies have mostly associated morbidity and mortality with ambient particle levels measured at one or a few central sites (e.g. Dockery et al. 1993, Pope et al. 2002), most likely due to the limited knowledge available on temporal and spatial variability of indoor particle concentrations and on indoor particle source emission strengths.

With no indoor sources present, ambient particles generated by industry, traffic and heating plants may penetrate the building envelope by ventilation and infiltration. In this situation, the outdoor particle concentration will determine the indoor concentration. Based on chemical mass balance modeling, Ward et al. (2006) estimated that wood combustion on average contributed to 82% of measured ambient PM2.5 in a particularly exposed valley. Hoek et al. (2008) assessed relationships between particle mass and particle number concentrations measured outdoors at a central site, right outside a home and inside the home. Altogether 152 homes with few indoor sources and located in Athens, Amsterdam, Helsinki, and Birmingham were included. The measurements were carried out during a week while the occupants were in their homes doing their daily whereabouts. The study found correlations between residential outdoor and indoor concentrations of particle mass in the ranges 0.35-0.85 (PM2.5), 0.52-0.72 (PM10), and 0.1-0.39 (PM10-PM25). Unfortunately, data from residences with and without wood stoves were not analyzed and compared separately.

Long et al. (2000) demonstrated that indoor fine particle concentrations in homes were significantly elevated during cooking, cleaning, and other general indoor activities involving combustion or physical movement. Indoor source events were typically of short duration, but could be very intense, especially for particles in the ultrafine and coarse modes. By analyzing time-activity information and concurrent data on particle concentrations in different size ranges recorded in three homes, Abt et al. (2000) confirmed that the relative contribution of outdoor and indoor particle sources to indoor concentrations varied considerably with the particle size. Indoor sources dominated the range from 2 – 10 μm and outdoor sources dominated the ultrafine range and fine modes.

Guo et al. (2008) studied the effect on indoor-outdoor correlations of particle number concentrations in an air conditioned classroom with

different air changes, the air change being the rate at which the indoor air volume is being replaced by outdoor air by ventilation and infiltration. Different ventilation scenarios were studied, including a natural ventilation mode with the air conditioning and the fans off and the windows open, which best may be compared with the conditions in most Danish homes. For this condition, Figure 1 shows the variation in the I/O ratio as a function of the outdoor particle concentration. Only rarely was the I/O ratio above 1 indicating that even with the windows open and in the absence of indoor sources, the indoor particle concentration was lower than the outdoor level. Nevertheless, indoor particle sources may increase indoor particle concentrations above the ambient level resulting in a higher I/O ratio than illustrated in Figure 1. The other tested scenarios with mechanical ventilation resulted in even lower I/O ratios.

Based on measurements carried out in a boarding school environment, Kingham et al. (2008) found that indoor PM1 and PM2.5 concentrations correlated well with outdoor concentrations and that indoor activity might resuspend coarser fractions of PM in the range PM2.5 to PM10. In their study, therefore, personal exposure to fine particles was mostly related to outdoor air pollution, which resulted primarily from heating of homes, irrespective of the person being inside or outside. Exposure to coarser particles was typically the result of resuspension caused by indoor activities.

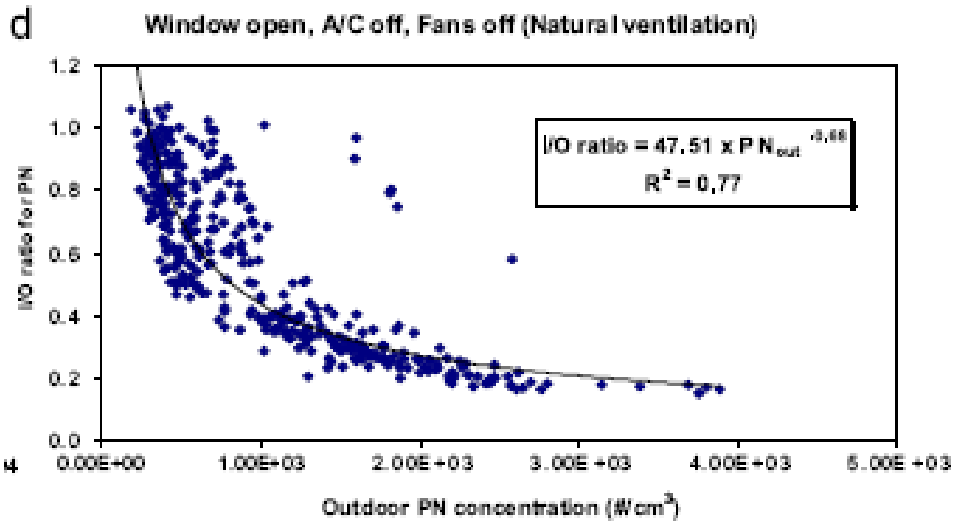

Figure 1. Indoor-outdoor particle concentration ratio versus outdoor particle number concentration with a natural ventilation scenario (From Guo et al. 2008).

2.2 Indoor particle sources

Many different indoor particle sources and their emissions have been identified and to some extent quantified, the most significant sources including tobacco smoking, heating, and cooking (Afshari et al. 2005, He

et al. 2004). Other causes of elevated indoor particle levels include resuspension by human or pet activities, cleaning, chemical processes in indoor air, and other processes related with human activities indoors (Long et al. 2000, Thatcher and Layton 1995, Abt et al. 2000).

Based on a laboratory study, Afshari et al. (2005) characterized selected indoor sources of fine and ultrafine particles, including candles, smoking and human activities. The highest concentration of particles in terms of numbers was observed with pure wax candles, although during burning predominantly in the ultrafine range. However, when the candles were extinguished, the concentration of larger particles peaked. Figure 2 shows the measured particle concentration profile with burning of pure wax candles.

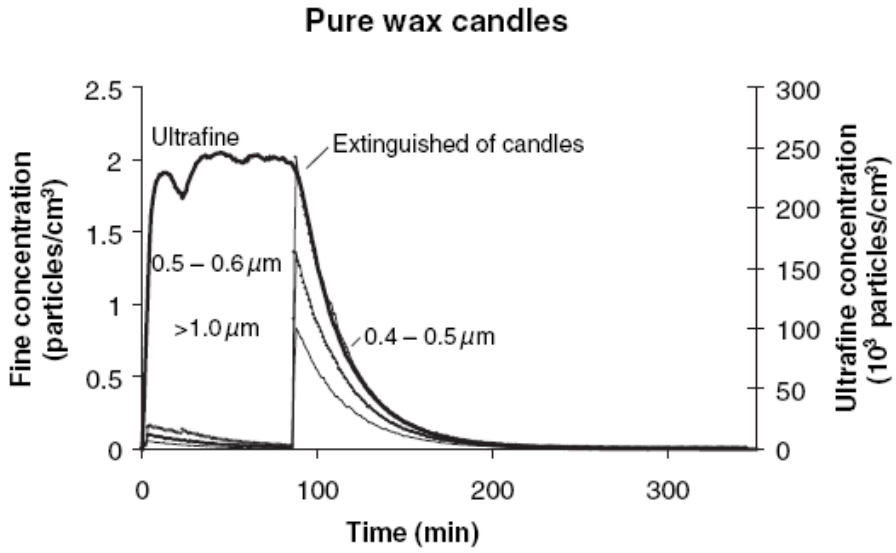

Figure 2. Concentration profile measured with burning of pure wax candles (From Afshari et al. 2005).

Afshari et al. (2005) also estimated the strengths of a range of other typical indoor particle sources. The most significant sources were heated radiator, heated electric stove, and frying of meat, the former two being temporary sources due to short-term aerosolization or resuspension of material sedimented on the surface of the heaters. In a field study in residential buildings located in Brisbane, He et al. (2004) identified the main indoor sources of particles and estimated their emission strength. Table 1, which is adopted from He et al. (2004), summarizes for PM2.5 and ultrafine particle number concentrations peak values, ratios of peak-to-background values and emission rates measured during 48 hrs in the residences. In correspondence with Afshari et al. (2005), cooking activities, in addition to smoking and cleaning, significantly contributed to elevate the indoor particle level above the baseline levels. Thus, the outdoor particle concentration may govern the indoor concentration during some periods, but human activities indoors in residential settings, especially related with heating or preparation of food, inevitably will affect the particle concentration. The location and duration of, and thus

the exposure to, elevated particle concentrations will depend on the available means of ventilation (e.g. a kitchen hood), on the outdoor air exchange rate, the between rooms airflows, and the type and location of the particle generation event.

Activity	N	Peak values ($\mu g\,m^{-3}$)		Ratio (mass)		Emission rate ($mg\,min^{-1}$)		Peak values ($particle\,cm^{-3} \times 10^3$)		Ratio (number)		Emission rate ($particle\,min^{-1} \times 10^{11}$)	
		Median	S.D.	Median	S.D.	Median	S.D.	Median	S.D.	Median	S.D.	Median	S.D.
Food preparation													
Cooking	24	37	194	2.89	12.6	0.11	0.99	126	177	10.3	19.3	5.67	8.61
Cooking pizza[a]	1	735		73.5		1.59		137.3		9.81		1.65	
Frying	4	745	352	33.6	28.3	2.68	2.18	154	21.3	10.0	6.1	4.75	2.45
Grilling	6	718	3427	90.1	312	2.78	17.8	161	69.9	8.69	5.27	7.34	5.06
Kettle	25	13	20	1.13	0.67	0.03	0.31	15.6	14.0	1.08	0.6	0.35	1.76
Microwave	18	16	18	1.12	0.42	0.03	0.11	16.3	28.6	1.12	1.55	0.55	1.94
Ovèn	6	24	6	1.76	0.53	0.03	0.03	61.5	31.9	2.96	0.78	1.27	2.10
Stove	4	57	264	2.4	19.7	0.24	1.29	179	287	12.5	10.5	7.33	51.4
Toasting	18	35	32	2.08	8.31	0.11	0.37	114	160	6.34	7.44	6.75	16.7
Other activities (multiple events)													
Open door	9	21	9	1.23	0.37			22.0	14.6	2.89	1.21		
Smoking	6	79	29	4.03	1.76	0.99	0.81	26.6	13.6	1.54	0.96	1.91	1.92
Sweep floor	3	35	4	2.04	1.3	0.05	0.01	34.9	5.86	1.05	0.01	0.12	0.02
Vacuuming	5	16	8	1.46	0.32	0.07	0.04	41.3	17.6	1.51	1.17	0.97	1.57
Washing	17	18	12	1.25	0.57	0.04	0.04	30.9	18.5	1.30	0.83	0.96	2.60
Other activities (single event)													
CVE oil[a]	1	132		13.2		0.91		74.6		8.29		5.52	
Dusting[a]	1	22		1.69		0.09		14.1		1.00			
Fan[a]	1	20		1.67				11.0		1.00			
Fan heater[a]	1	15		1.50		0.05		87.1		27.2		4.07	
Hair dryer[a]	1	45		1.36		0.04		9.5		1.06		0.11	
Shower[a]	1	20		1.08		0.04		10.7		1.37		0.78	
Washing M[a]	1	43		2.05		0.12		11.1		1.18		0.15	

Note: N: sample number; CVE oil: candle vapour eucalypt oil; Washing M: washing machine.
[a]no S.D.

Table 1. Summary of the results of 48 hr measurement of PM2.5 and ultrafine particle numbers in residences: peak concentration values, ratio of peak to background concentration and estimated emission rates (From He et al. 2004).

3 Indoor-outdoor measurements

The Slagslunde measuring campaign took place in December 2006 - March 2007 in the village of Slagslunde ca. 25 km North of Copenhagen. The campaign comprised measurements of many air pollution components at two sites: an exposed site within the village, and a background site outside of it.

Indoor and outdoor pollution was measured simultaneously for one week in two houses: one with a wood stove (March 6-12), and another without a stove (March 12-18). The wood stove was a Lotus stove from 1997. Both houses were located close to the measuring site in the village

(at a distance of 20 m for the house with wood stove, and 50 m for the house without). Focus was on pollution with soot, because soot is more specific for wood smoke than PM2.5. PM2.5 can be caused by many other activities than wood combustion. Soot measurements were performed with Particulate Soot Absorption Photometers (PSAP) with a time resolution of 15 minutes as described by Wåhlin et al. (2010). The residents of the houses logged activities such as cooking, cleaning, opening windows etc.

Using a passive tracer gas technique (PFT method), integrated measures of outdoor to indoor transport of air as well as between living room and bedroom and between bedroom and living room in each residence was measured simultaneously with the soot measurement (Bergsøe 1992). Two identical sources constantly emitting tracer gas of one type were set up in the living room and two other sources in the bedroom (or in a bedroom and office in the residence without a woodstove, representing the second zone in that residence). Also, two adsorption tubes were set up in each room. After one week exposure in each residence, the adsorption tubes were analyzed and yielded information on outdoor air supply and airflow between the rooms, where the occupants were expected to spend most of their time while occupying the house. Figure 3 shows schematically the two residences and the location of the wood stove and the emission and adsorption tubes.

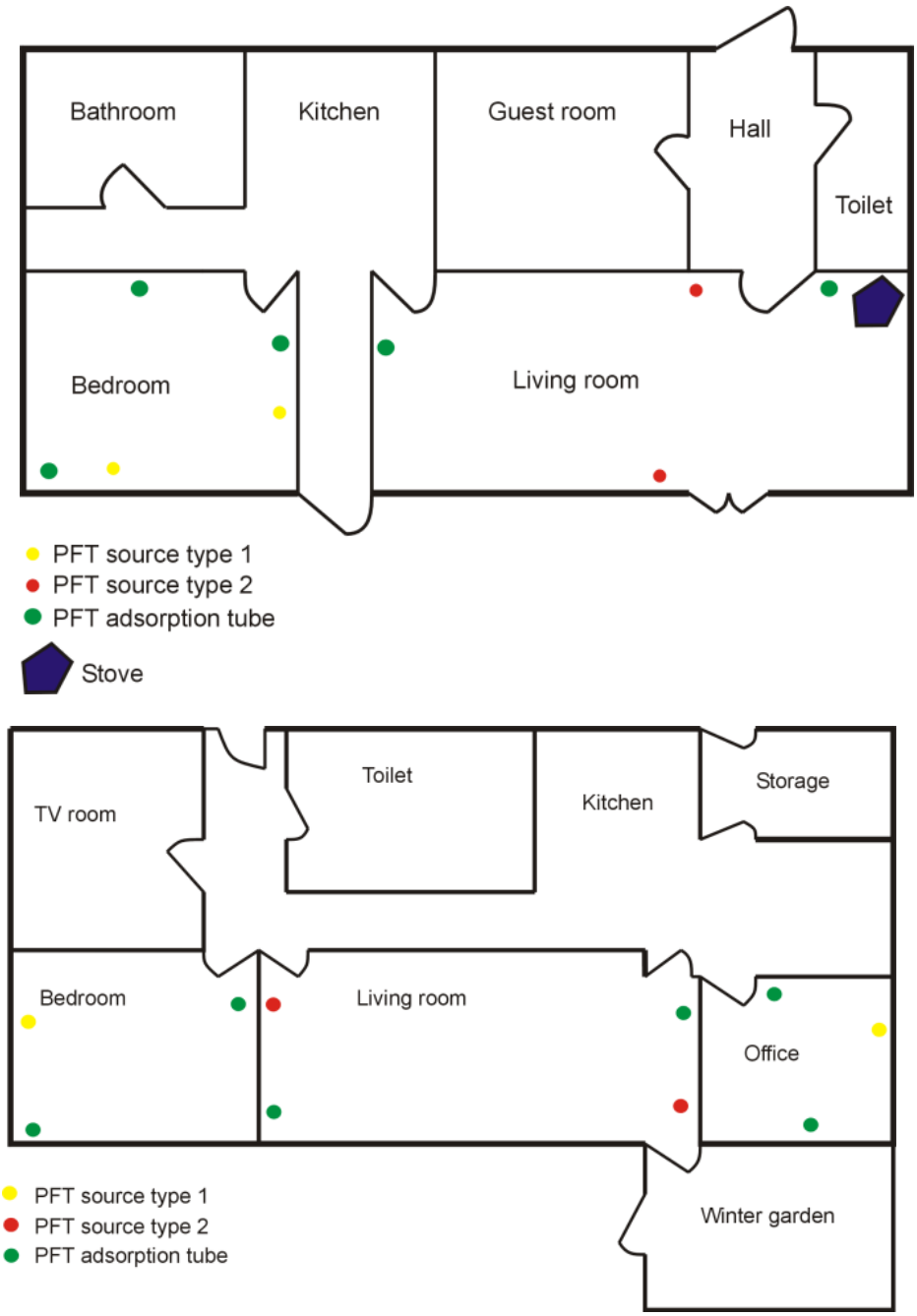

Figure 3. Schematic of the residences. Top: residence with a wood stove. Bottom: residence with no woodstove.

3.1 Results

Table 2 summarizes the airflows determined from the passive tracer gas measurements.

	Outdoor air supply		Inter-zonal airflow		Average air-change in residence (h^{-1})
	Living room (m^3/h)	Bedroom / office (m^3/h)	Living room to bedroom (m^3/h)	Bedroom to living room (m^3/h)	
Residence with wood stove	32	31	35	62	0.6
Residence without wood stove	24	35	122	204	0.5

Table 2. Summary of the airflow rates measured in each residence during two succeeding weeks.

In the residence without a wood stove, the airflow rates between the living room and the adjacent rooms were too high to divide the house into several zones, and particle concentrations would therefore exhibit smaller variation in this than in the other visited house, where division of the bedroom and the living room would be meaningful.

Figure 4 shows the indoor-outdoor soot ratio measured in the two residences. Especially in the residence with a wood stove, the I/O ratio peaked almost daily. The activity pattern form completed by the residents revealed that the peaks occurred synchronously with the start of a wood stove heating period, i.e. when the doors to the stove were open and the chimney not being warmed up, resulting in initially negligible buoyancy and thus dispersion of particles in the living room. In the residence without a wood stove the I/O ratio peaked only a few times, typically as a result of toasting bread.

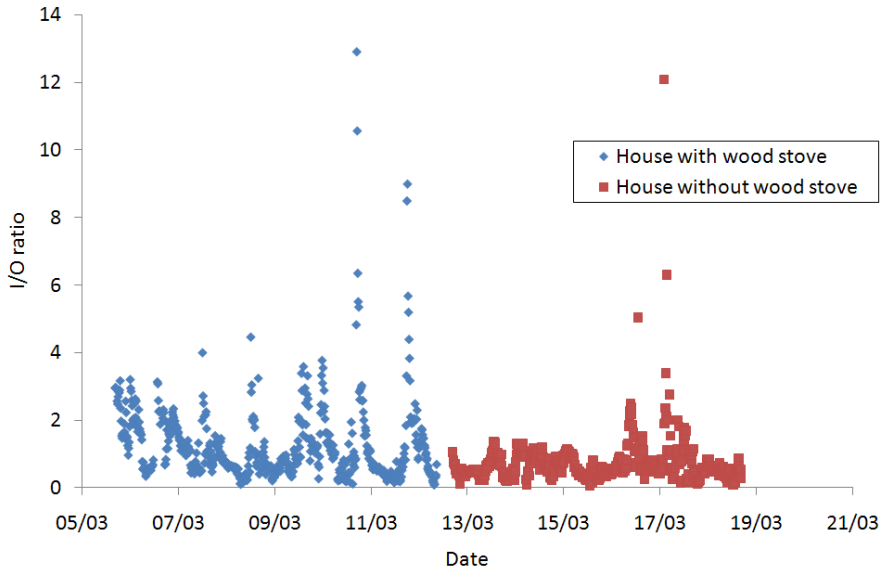

Figure 4. Indoor-outdoor ratio of the concentration of soot measured in the residences with and without a wood stove during two consecutive weeks.

Figure 5 confirms that the I/O ratio appeared to be higher in the residence with the wood stove. The difference was significant as determined by the outcome of a t-test carried out on log-transformed I/O ratio data obtained in each residence (unpaired, one-tailed t-test, $p < 0.001$). In addition, the average outdoor concentration measured in the village was significantly higher (4.8 vs. 4.4 μm^{-1}) during the second period (unpaired, one-tailed t-test, $p < 0.001$), when measurements were carried out in the residence without a wood stove, indicating that it was the indoor sources that caused higher exposure to wood stove generated particles in the residence with the wood stove.

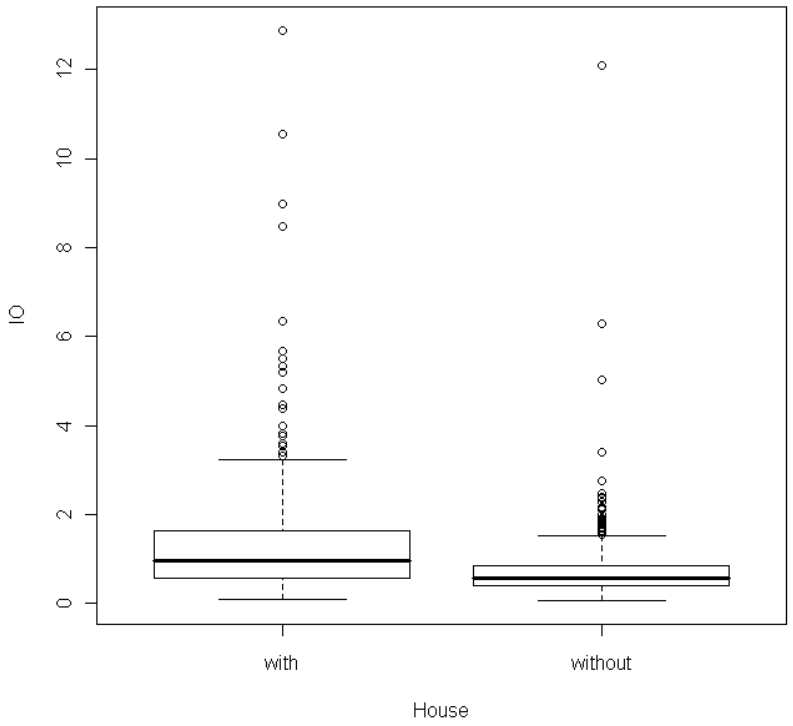

Figure 5. Box plot of soot I/O ratio measured in the residence with and without a wood stove. The bold, horisontal line indicates the median, the box the inter-quartile range, and the whiskers extend to the most extreme data point which is no more than 1.5 times the height of the box away from the box.

In homes without indoor particle sources correlation between ambient and indoor particle concentration may be meaningful, but the same may not hold true with many or strong indoor sources. This is confirmed by Table 3, which compares Pearson correlation calculated for the ambient background, village background and the indoor soot concentration measured at 15 min intervals during the one-week measurement period in the two residences.

	Residence with wood stove			Residence without wood stove		
	Indoor	Village	Background	Indoor	Village	Background
Indoor	1	0.22	0.31	1	0.74	0.83
Village	-	1	0.40	-	1	0.84
Background	-	-	1	-	-	1

Table 3. Pearson correlation coefficient between instantaneous values of residential-village, residential-background, and village-background soot measured in the two residences every 15 min during one week.

Clearly, correlations between indoor and ambient particle mass concentrations were higher in the residence without the wood stove. Naturally, the correlation will depend on occupant activities and the air exchange rate in the residence. The air exchange rate differed only little between the two residences, whereas occupant activities, in addition to the wood stove, differed between the residences.

4 Modeling of particle concentration in the residence bedroom

In most Danish residences with a wood stove, the stove is located in the living room, thus increasing the infiltration rate while the stove is in use and the risk of indoor particle emission when stove heating is initiated or when new fuel is added to the stove. While the exposure may be highest during occupancy of the living room, residents may stay longer in other rooms, e.g. the bedroom, which may be considered another zone in the residence where the particle exposure is affected by the conditions in the living room. In this section, the time-averaged ventilation data presented in Table 2 together with the measured living room particle concentrations were used to assess the particle exposure in the bedroom.

Several processes affect the concentration and fate of indoor airborne particles, which are often described by mass-balance models, simplified to enable practical use. Nazaroff (2004) describes one such model, accounting for ventilation, infiltration, and indoor emissions. This model in a slightly modified form was used to assess particle exposure in the bedroom of the residence with a wood stove. Figure 6 shows schematically the mechanisms that govern the indoor particle concentrations in the residences (modified from Nazaroff 2004).

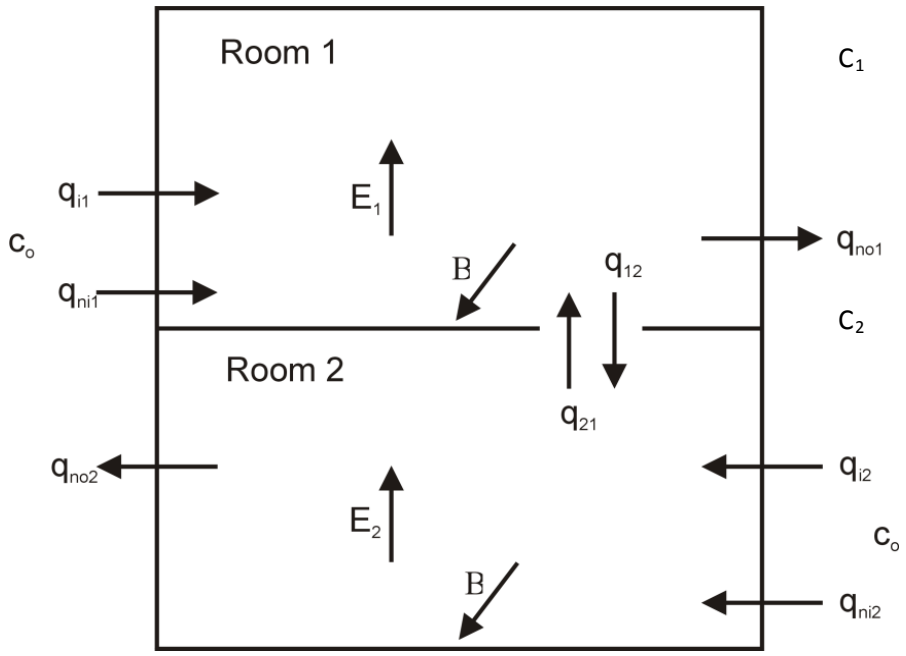

Figure 6. Schematic representation of the mechanisms governing the indoor particle concentration.

Outdoor air with a particle concentration of c_o enters the building via two possible flow pathways: natural ventilation q_n; and infiltration through leaks and cracks q_i (mechanical supply of ventilation air was considered irrelevant in the two residences). Different inter-zonal airflows from adjacent rooms with a concentration of c_{ji} and flow rate q_{ji}, and to adjacent rooms at a flow rate of q_{ij} and concentration c_i may also affect the concentration.

The natural ventilation flow is typically passing through relatively large openings, and it is assumed that there is no loss of particles. However, only a fraction of the particles in the infiltration airflow P are assumed to enter into the indoor environment.. Within the space, there may be emission of particles at the rate E and deposition onto surfaces at a rate that may be described by a first-order loss-rate coefficient β. Assuming that the particle concentration is uniform throughout the interior volume, conservation of mass may be used to formulate the governing equations for the indoor particle concentration:

$$V_1 \cdot \frac{dc_1}{dt} = \frac{(E_1 + c_o \cdot (q_{ni1} + P \cdot q_{i1}) + q_{21} \cdot c_2(t) - c_1(t) \cdot (q_{no1} + \beta \cdot V_1))}{V_1}$$

(eq. 1)

$$V_2 \cdot \frac{dc_2}{dt} = \frac{(E_2 + c_o \cdot (q_{ni2} + P \cdot q_{i2}) + q_{12} \cdot c_1(t) - c_2(t) \cdot (q_{no2} + \beta \cdot V_2))}{V_2}$$

(eq. 2)

In these coupled differential equations index 1 refers to the living room and index 2 to the bedroom. V is the room volume, c the time-dependent indoor concentration, c_o the outdoor concentration (considered constant

during the simulation period), q_{ni} the air supply through larger openings, q_i the infiltration airflow through leaks and cracks, P the fraction of particles that penetrate the building envelope through leaks and cracks, q_{ij} the airflow from room i to room j, q_{no} the airflow out of the room (sum of q_{ni} and q_i), and β the first order loss-rate coefficient.

It is assumed that pressure differences between spaces are rather modest, and airflow between rooms therefore is limited to large openings. Inter-zonal transfer of particles is therefore not attenuated such as in the flow through leaks and cracks, and particle transfer depends only on the airflow rate and the particle concentration in the room supplying the airflow.

The penetration efficiency P is affected by several factors, such as outdoor particle concentrations and meteorological conditions (Guo et al. 2008). However, Liu and Nazaroff (2001) modeled the penetration of particles through a crack in the building envelope and found that in the size range from <0.07 μm to >3 μm, the penetration coefficient was close to 1. Nazaroff (2004) also presents values for the deposition rate coefficient β which varied in the range 0.08 for particles around 0.2 μm up to 0.2 for a particle size of 0.02 μm. It is assumed that the size of particles generated by wood smoke is <0.2 μm and thus an estimated average value of β of 0.11 was used in the simulations.

The mathematical software Maple (Maple 2007) was used to solve numerically the coupled differential equations 1 and 2 after an event that generated a high particle concentration in the living room (start of wood stove heating). Input to the solution was the flow rates listed in Table 2, assumed negligible emission in both rooms after the event (decay of the particle concentration) and negligible flow through leaks and cracks as compared with the natural ventilation flow

Figure 7 illustrates an example of a simulation result and shows how the particle concentration in the bedroom may vary with the concentration profile obtained in the living room. It should be noted, however, that the bedroom particle concentration profile gives only a rather crude estimate of the exposure in the bedroom, since the flow rates were average values measured during a full week, and other input parameters based on somewhat loose assumptions. Nevertheless, Figure 7 shows that an event that generates a high particle concentration in one room may affect the concentration, and thus the exposure, in adjacent rooms several hours after the event, although with the recorded airflows the peak concentration in the bedroom was considerably lower than in the living room. More detailed information on the airflows in the residence would increase the validity of the simulations. In Figure 7, the event that generated the high particle concentration occurred already at 18:00 hrs (time 0 in the figure) and therefore the estimated concentration in the bedroom has returned to the background level at the occupants' presumed bedtime. However, events occurring later may continue to cause elevated exposure into the sleep period.

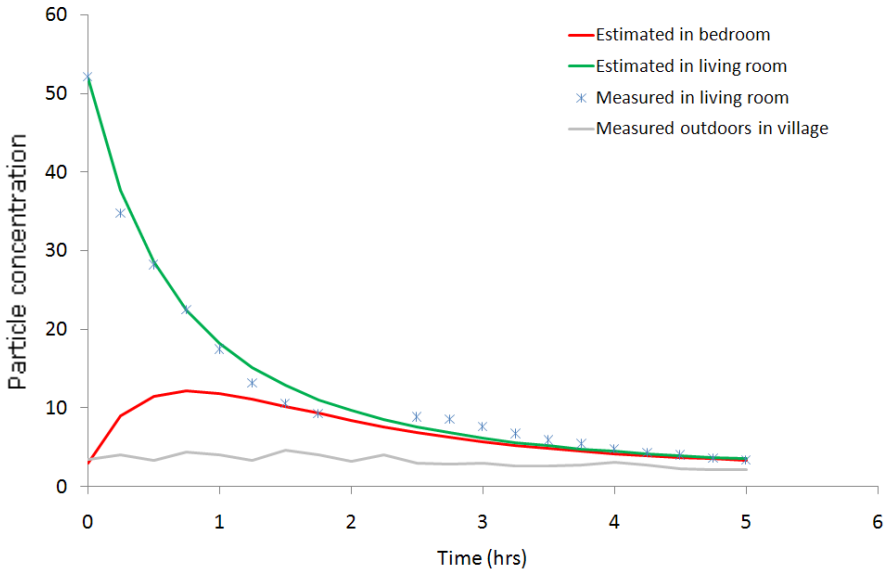

Figure 7. Result of a simulation of the particle exposure in the bedroom following a particle generating event in the living room.

In summary, the indoor-outdoor measurements showed that the occupants' use of the wood stove in one residence resulted in a significantly higher I/O ratio, and thus higher exposure to particles in that residence, than in the residence without a wood stove. The I/O ratio in the residence with a wood stove also fluctuated more and reached higher and more frequent extreme values than in the other residence indicating that use of a wood stove may be a strong and intermittent indoor source of particles. The events that generated high particle concentrations affected the concentration and thus the exposure in adjacent rooms during several hours after the event. These findings were based on measurements in a limited number of residences, on a combination of time-averaged and time-resolved data, and on several other assumptions, and therefore should be substantiated by additional experimental evidence.

Particle generation indoors may be caused by many other sources than wood stoves (e.g. Table 1). Most significant are activities that include combustion, e.g. food preparation and burning of candles, although the latter primarily in the ultrafine mode. Dispersion of particles that follow from preparation of food is typically limited to some extent by an exhaust hood in homes in industrialized countries, but burning of candles is a leisurely activity that may elevate particle concentrations in several rooms in a residence and often during longer periods. Quantification of wood stove particle emission rates are scarce in the literature, but as indicated in this study, wood burning in a stove when it is started may yield high indoor particle concentrations. However, after some time, the stove will increase the air exchange rate in the residence and contribute to reduce the particle concentration provided the outdoor concentration is lower than indoors.

Relevant to the current study is the newly reported findings of Ward et al. (2008) who measured PM2.5, levoglucosan and other markers of wood combustion in 16 homes in a U.S. town before and after replacing old woodstoves with new, EPA-certified woodstoves. The 24 h average indoor PM2.5 decreased significantly (71%) before and after replacing the old stove (51.2 ± 32 μg/m^3 (median 34.5 μg/m^3) vs. 15.0 ± 20.8 μg/m^3 (median 9.5 μg/m^3)). Similarly the maximum PM2.5 was reduced from 434 ± 419 μg/m3 (median 266 μg/m^3) to 103 ± 167 μg/m^3 (median 51.5 μg/m^3). Ambient PM2.5 was not significantly associated with indoor PM2.5 or indoor levoglucosan, nor did ambient PM2.5 measures have significant interactions with woodstove changeout for changes in indoor PM or levoglucosan. The results indicate that exposure indoors to combustion products may be reduced by the use of newer stoves that abide by current emission guidelines.

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